

# Cavitation-induced force transition in confined viscous liquids under traction

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We perform traction experiments on simple liquids highly confined between parallel plates. At small separation rates, we observe a simple response corresponding to a convergent Poiseuille flow. Dramatic changes in the force response occur at high separation rates, with the appearance of a force plateau followed by an abrupt drop. By direct observation in the course of the experiment, we show that cavitation accounts for these features which are reminiscent of the utmost complex behavior of adhesive films under traction. Surprisingly enough, this is observed here in purely viscous fluids.

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Adhesive materials are often tested by way of the so-called probe-tack test [1] which tends to mimic the detachment of an adhesive joint. The material is used as a thin film (typically  $100\mu\text{m}$  in thickness) deposited on a flat, rigid substrate. A flat, solid punch (also called indenter or probe) is approached and kept in contact with the film for a few seconds. It is then pulled away at some prescribed, constant velocity, while the applied force is recorded. Good adhesives typically yield a force response with the following characteristic features [2, 3]: the force increases sharply and reaches a peak, followed by a plateau at a lower value, until it drops quite abruptly to much lower values and finally vanishes when complete separation is achieved. In recent years, direct observation of the film during traction has been conducted [4]. Meniscus instabilities [5, 6], bubble growth and fibril formation are commonly observed. Some of these phenomena have been associated to particular features of the force response: force peak for the bubble appearance [4] and force plateau for the bubble growth [4] or fibril elongation [1]. Such behaviors have been attributed to the specific rheological properties of adhesive materials. Otherwise, many studies have been devoted to the instabilities of viscous liquids confined between two parallel plates [7] under traction, a geometry sometimes named “lifted Hele-Shaw cell”. During the traction, the fluid has to flow inwards. Due to the resulting pressure gradient, the edge of the sample destabilizes from its initial, regular shape through the Saffman–Taylor mechanism [8, 9]. As these instabilities develop, air fingers grow towards the center of the sample, producing characteristic fingering patterns [10]. At the end of the traction process, interesting instabilities occur prior to the detachment of the liquid column [11].

In the present Letter, we study the force response of viscous liquids in combination with pattern observations. Our aim is to determine which phenomena observed in adhesive materials rely on their specific properties and which of them are more general. We show indeed that the force plateau and subsequent drop observed in adhesives are also present in viscous liquids and that the

mechanisms involved are similar.

The apparatus consists in two plane, horizontal plates whose separation can be varied. A drop of liquid is initially deposited on the bottom plate. The glass top plate is slowly approached until the drop is confined into a film of prescribed thickness. The top plate is then pulled at constant velocity  $V$  (in the range  $1\mu\text{m/s} - 1\text{mm/s}$ ) while the force is being recorded *via* a transducer.

Following the mechano-optical design described in Ref. [12], we observe the liquid–glass interface through a built-in internally-reflecting prism used as the top plate [13]. In addition, a picture is made of the patterns left in the liquid on the lower plate after full separation is achieved.

The liquid used is a highly viscous, non-volatile silicon oil (Rhodia 47V1000000, viscosity  $\eta = 10^3\text{Pa.s}$ ). In all experiments, the sample volume is kept constant. Initially, the thickness is  $h_0 = 100\mu\text{m}$  and the diameter of the squeezed drop is  $9\text{mm}$ . The oil has been left in a vacuum chamber (pressure  $10^{-4}\text{atm}$ ) for one hour in order to remove entrapped gas bubbles and achieve a reproducible initial sample state.

We plot the force versus the plate separation in a log-log representation. The separation is obtained by subtracting the machine elongation from the nominal separation [15]. The machine rigidity  $K = 7.5 \cdot 10^5\text{N/m}$  has been measured separately.

Force-displacement curves obtained at different traction velocities are shown in Fig. 1. Two different types of behaviors for the force decrease are clearly observed. The force decrease is smooth at low velocities. By contrast, a force plateau immediately followed by a sharp drop appears at high velocities, a feature strikingly reminiscent of adhesives. Moreover, a weak noise, resembling the “pop” produced by the opening of a bottle of wine, is heard distinctly in this case. The transition between these two behaviors can be assigned to the first curve that displays an inflection point, namely  $V_c \simeq 15\mu\text{m/s}$ . Fig. 2 shows pictures of the glass–liquid interface taken during

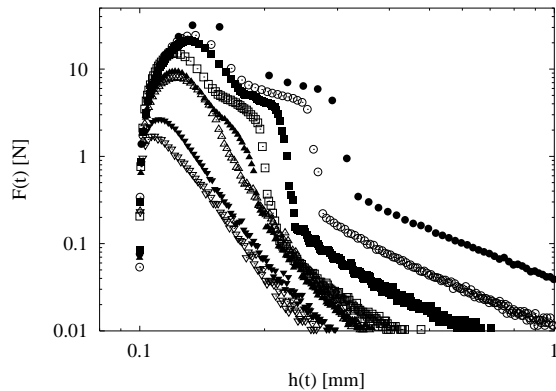


FIG. 1: Force versus plate displacement at separation rates ranging from  $V = 1\mu\text{m/s}$  ( $\nabla$ ) to  $V = 1\text{mm/s}$  ( $\bullet$ ). The force plateau and drop appear only above some critical velocity ( $\blacktriangle$ ,  $V_c = 15\mu\text{m/s}$ ).

the traction and the associated force curves (lin-log representation), for three different velocities. At low velocity (Fig. 2a,  $V < V_c$ ), Saffman–Taylor instabilities develop and the resulting progressive finger growth corresponds to the smooth decrease of the force. At high velocity (Fig. 2c,  $V > V_c$ ), cavitation is observed together with the force plateau. More precisely, the pictures show that numerous bubbles appear just before the force peak and that they grow and develop in the course of the plateau. Thus, in purely viscous liquids and in adhesives alike, the existence of a plateau is associated to cavitation [4]. At  $V \simeq V_c$  (Fig. 2b), one clearly sees the occurrence of both fingering and cavitation together, with the growth of a single bubble and its subsequent collapse. This is confirmed by pictures of the sample taken immediately after full separation (Fig. 3): only above  $V_c \simeq 15\mu\text{m/s}$  (Fig. 3, top right) do cells appear in the center of the sample, suggesting that cavitation has taken place.

From these observations, we can suggest an interpretation of what happens during the traction over the whole velocity range. The general idea is that as the plates are being pulled apart, volume conservation *a priori* implies the existence of a radial, convergent flow from the edge of the sample towards the center, which tends to relieve the stress. Fingering instabilities and cavitation compete in further relieving the applied stress. The reason why fingering appears at low velocities while cavitation takes over at high velocities is the lower threshold force for fingering and the higher bubble growth rate for cavitation [16, 17].

Let us now focus on high velocities and in particular on the unexpected features of the force response: the plateau and the subsequent force drop. As described above, the force plateau corresponds to the bubble growth. The explanation goes as follows. The pressure recorded on

the plate is the difference between the atmospheric pressure and the pressure in the sample. In the presence of bubbles, the pressure in the sample is the sum of a contribution from the pressure inside the bubbles and a contribution (which is small in the present situation) from the flow in the liquid that surrounds the growing bubbles [18]. The pressure inside the bubbles results from two possible sources. (i) If gas was present initially in the bubbles, the tremendous volume increase has made its contribution to the pressure to practically vanish. (ii) If the liquid is somewhat volatile or contains a volatile component, the vapor phase may contribute a pressure whose value cannot exceed the saturating vapor pressure  $p_{sat}$ , in which case, the plateau corresponds to a liquid–vapor phase transition. In both cases, the bubble pressure contribution is essentially constant.

At some point of the bubble growth, a few liquid walls between bubbles break, thus allowing air from the outside to suddenly rush into the sample from the edge. This produces the observed abrupt drop of the force, since the interior of the former bubbles now communicates with the outside air, thus relieving the pressure difference. This is also responsible for the “pop” which can be heard distinctly as mentioned above. The total pressure on the plate is thus expected to drop by  $p_{atm} - p_{sat}$  at the end of the force plateau. The curves in Fig. 1 show that the force drop at the end of the plateau is on the order of 5 N, for all separation rates  $V$  in the high velocity regime ( $V \geq V_c$ ). Since the sample diameter is 9 mm, this corresponds to a pressure of about 1 atm.

The pressure difference is now relieved, and there remains the sole contribution from the liquid walls. From basic hydrodynamics [19], the flow is elongational and the force can be expressed as:

$$F(t) = \frac{4\eta\Omega V}{h^2(t)} \quad (1)$$

where  $\Omega$  is the total volume of the walls and  $h(t)$  the plate separation at time  $t$ . Fig. 4, drawn without any adjustable parameters, shows that the estimate of the force is quite good over a velocity range of almost two orders of magnitude.

Let us now turn to the low velocity regime. There is no cavitation. If we neglect fingering instabilities, the flow at early times is radial and convergent, and locally parabolic (Poiseuille flow) [19]. The force exerted on the plates is then:

$$F(t) = \frac{3}{2\pi} \frac{\eta\Omega^2 V}{h^5(t)} \quad (2)$$

Fig. 5 shows that despite the fingering, this law is quantitatively verified at early times for low separation rates, once again without any adjustable parameters. At slightly later times, *i.e.*, as fingers have sufficiently developed, thus relieving the stress, the real force decreases

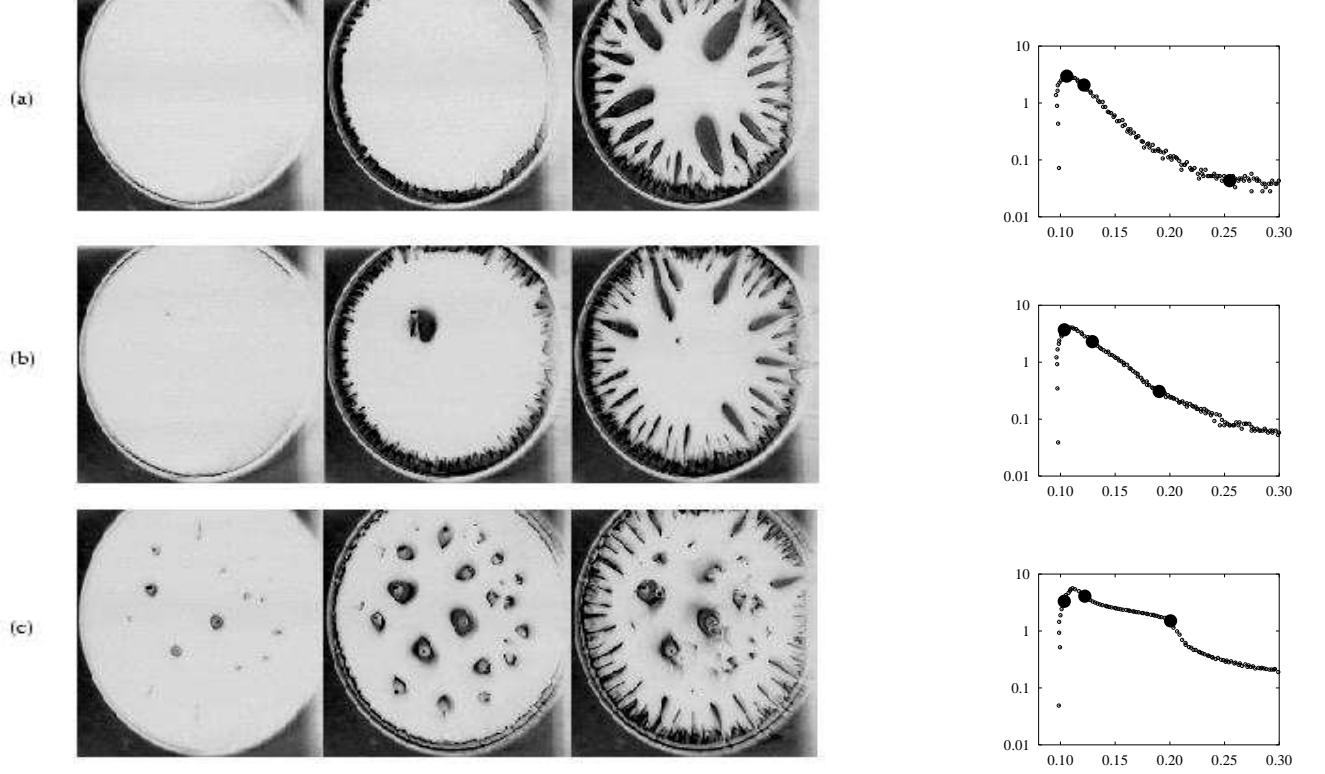


FIG. 2: Instantaneous pictures of the liquid-glass interface during the traction experiment at (a)  $V = 8 \mu\text{m/s} < V_c$ , (b)  $V = 16 \mu\text{m/s} \approx V_c$  and (c)  $V = 100 \mu\text{m/s} > V_c$ , as revealed through a built-in internally-reflecting prism used as the top plate. Inverted gray scale: white regions are wet by the fluid and do not reflect light; the glass surface is in contact with vapor in dark regions. The corresponding force-separation curves are displayed, with filled circles indicating when the images were recorded.

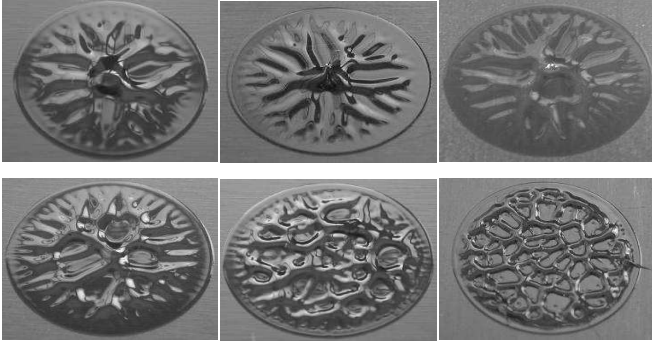


FIG. 3: Patterns left on the bottom plate immediately after full separation was achieved (increasing velocities from left to right and from top to bottom; top right is  $V = V_c$ ). Note the arborescent pattern [10] at low velocities and the cellular pattern for  $V > V_c$ .

somewhat more rapidly. At even later times, we might expect equation (1) to be valid ( $F \propto h^{-2}V$ ). However, at such low velocities, the corresponding force lies within the experimental resolution and is, furthermore, masked by capillarity [16].

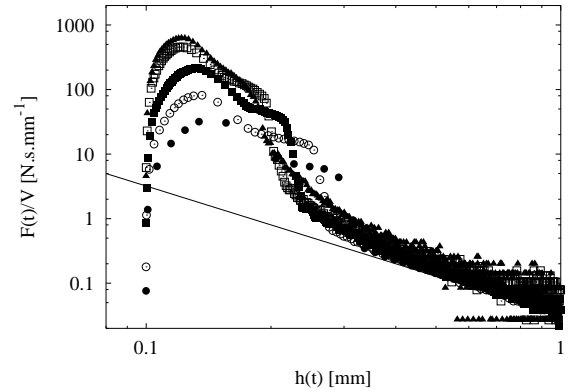


FIG. 4: Force (normalized by the traction velocity) versus plate displacement for  $V$  ranging from  $V = V_c = 15 \mu\text{m/s}$  ( $\blacktriangle$ ) to  $V = 1000 \mu\text{m/s}$  ( $\bullet$ ). The straight line corresponds to equation (1) without any adjustable parameters.

Let us now compare the behavior of purely viscous liquids to adhesives. The mechanisms related to the plateau and the subsequent force drop are similar [4, 20], even though the plateau is generally much higher and much longer in adhesives. The height of the plateau is ex-

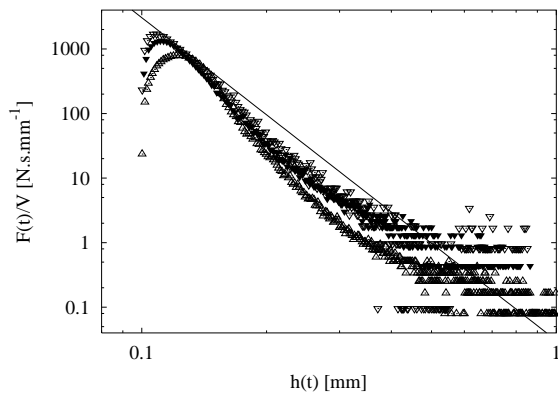


FIG. 5: Force (normalized by the traction velocity) versus plate displacement for  $V < V_c$ . The straight line corresponds to equation (2) without any adjustable parameters.

plained by the contribution from the viscoelastic flow, which is not negligible any more compared to atmospheric pressure. The walls are also more resistant to air penetration, explaining the length of the plateau. As for the force drop, all these observations and interpretations lead us to believe that whatever the material, a sudden air penetration in the sample produces a pressure drop of 1 atm.

By conducting traction experiments on confined, purely viscous liquids, we have observed and interpreted new behaviors, namely cavitation and its competition with fingering instabilities. A remarkable feature of the force response is the appearance of a plateau at high velocities as observed in adhesives. It can be interpreted in terms of the liquid being eventually full of almost empty bubbles. This shows that the bubbles, the force peak and the plateau are far from specific to adhesive materials: only the relative importance of two contributions to the plateau force differs. The appearance of a force plateau for simple liquids is thus interesting not only *per se*, but also in that it brings new elements to understand what happens in adhesive materials.

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